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Large Induced Magnetic Anisotropy in Pulsed Laser Deposited Manganese Spinel Ferrite Films

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Here, we report the oxygen-pressure dependence of magnetic anisotropy in pulse laser deposited MnFe_2O_4 films. Magnetic anisotropy fields are shown to reach 5 kOe when the films are processed at oxygen pressures below 5 mTorr. The bulk values are around 200 Oe. Further, the preferred direction of magnetization can be aligned either along the film plane ($p_{\text{ox}} < 8$ mTorr), as is typical, or perpendicular to it ($p_{\text{ox}} > 8$ mTorr). The ability to induce large perpendicular magnetic anisotropy in spinel ferrites allows for new applications above S-band frequencies (i.e. phase shifters, filters, isolators, and circulators) to be considered.

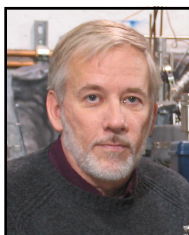
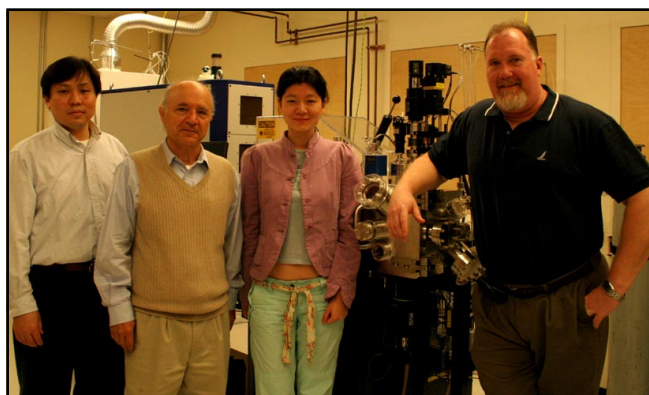
Manganese ferrite (MnFe_2O_4) is a well-studied spinel ferrite that has low magnetic anisotropy (H_a) at room temperature ($K_1 = -33 \times 10^3$ erg/cm³ or $H_u = 2|K_1|/M_s = 175$ Oe) arising from the low magnetocrystalline anisotropy energy common to cubic structures. This low H_u value limits the applications of this ferrite, or more generally, most cubic spinel ferrites, to frequencies at or below the S-band (1-5 GHz). To overcome this limitation, large magnetic fields are needed to achieve a high ferromagnetic resonance frequency and increase the bandwidth of these materials. These large magnetic fields do not allow for the miniaturization of such potential devices, however.

The enhancement of the magnetic anisotropy in manganese ferrite is related to the oxygen processing pressure used in the pulsed laser deposition process (**Figure 1**). We measure a

large growth-induced anisotropy for both low ($H_u \sim 5000$ Oe) and high oxygen-processing pressures (~ 1500 Oe). The natures of these magnetic anisotropies are quite different. For example, at low oxygen-processing pressures the magnetization aligns in the film

plane, as one might expect from the influence of the demagnetizing energy. However, at high pressures the anisotropy aligns perpendicular to the film plane.

One possible source of this magnetic anisotropy derives from the distribution of magnetic cations within the unit cell. In order to measure the cation distribution, select samples were subjected to EXAFS (extended x-ray absorption fine structure) measurements. Data collection was performed using beamline X23B at the National Synchrotron Light Source in fluorescence yield at room temperature. At the time data were collected, the storage-ring energy was 2.54 GeV and the ring current ranged from 180-250mA. Here, we applied a multi-edge refinement using Athena and Artemis codes of Ravel and Newville, respectively, to analyze the cation distribution of these samples,



Authors (top from left) Soack Dae Yoon, Carmine Vittoria, Aria Yang, and Vince Harris (bottom from left) Johnny Kirkland, Xu Zuo, and Joseph Christodoulides.

which were produced under different oxygen pressures. **Figure 2** is a plot of representative best-fit data for both Mn and Fe EXAFS.

With an increase in oxygen pressure used in processing, the octahedral site occupancy of the manganese ions increases, as measured using EXAFS, from 32% to greater than 50%. This is compared with the bulk equilibrium distribution of 20%. Correspondingly, the Fe octahedral occupation decreases from 84% to 75%. The difference in site distribution may be influenced by the difference in cation valence caused by the oxygen pressure, lattice strain from the mismatch in lattice constants and thermal expansion coefficients, or from the large effective quench rate.

As the oxygen pressure used in processing is increased from 1 to 50 mTorr there are significant changes in the physical state of the films. We envision that at low oxygen pressures ($p < 5$ mTorr), anion defects arise from the incomplete oxidation of cations on the surface of the growing film. These defects lead to a greater occupation of Fe^{2+} on the octahedral sites, providing large contributions to the uniaxial anisotropy constant from a single ion anisotropy mechanism. At pressures greater than 8 mTorr, H_u becomes negative and the magnetization aligns perpendicular to the film plane. At higher pressures the ions in the ablated flux experience more collisions en route to the substrate, which reduces their kinetic energy and

subsequently reduces their mobility on the surface of the growing film. This lack of mobility leads to the freezing in of cation disorder. In the case of MnFe_2O_4 this results in an increase in the inversion of Mn cations, that is, the distribution of Mn on the octahedral sublattice. Another interesting trend seen in the EXAFS analysis is the increase in the oxygen displacement parameter (commonly denoted by u). We speculate that this distortion is a direct consequence of the local strain resulting from the cation disorder and results in a tetragonal distortion of the unit cell. This breaking of the crystal symmetry leads to perpendicular anisotropy via a magnetocrystalline anisotropy mechanism.

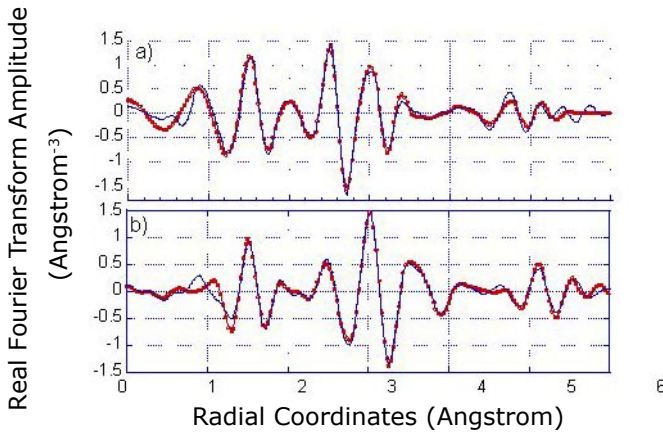


Figure 1. EXAFS data as the real part of the Fourier transform from Mn (a) and Fe (b) K-edge absorption (solid curve) with best fit data (symbols) for a MnFe_2O_4 film deposited on a (100) MgO substrate at an oxygen pressure of 1 mTorr.

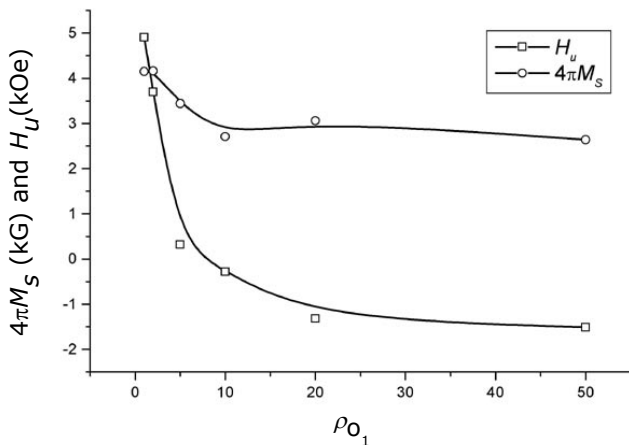


Figure 2. Uniaxial magnetic anisotropy field (H_u) and saturation magnetization ($4\pi M_s$) as functions of oxygen pressure used in PLD growth. M_e is the vector defining the magnetic easy axis.